FINAL REPORT

Task 5: Recommendations for Monitoring to Verify Phases I and II – Part 1

November 1994

Submitted to the Colorado Department of Public Health and Environment, Disease Control and Environmental Epidemiology Division, Rocky Flats Health Studies in partial fulfillment of Contract No. 100APPRCODE 391

"Setting the standard in environmental health"

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ACRONYMS

CDPHE	Colorado Department of Public Health and Environment
CESC	Citizens Environmental Sampling Committee
CSU	Colorado State University
DOE	U.S. Department of Energy
EG&G	EG&G Rocky Flats, Inc.
HAP	(Rocky Flats) Health Advisory Panel
QA	quality assurance
QC	quality control
RAC	*Radiological Assessments Corporation*[1]
TTLα	total long-lived alpha
VOC	volatile organic compound

[1] In 1998 *Radiological Assessments Corporation* changed its name to *Risk Assessment Corporation*. For consistency throughout the project, all reports were published by *Radiological Assessments Corporation*.

TASK 5: RECOMMENDATIONS FOR MONITORING TO VERIFY PHASES I AND II-PART 1
FINAL REPORT

INTRODUCTION

The Rocky Flats Environmental Technology Site is owned by the U.S. Department of Energy (DOE) and is contractor-operated by EG&G Rocky Flats, Inc. (EG&G), and until recently was a nuclear weapons research, development, and production complex. The RFP is located on 6,550 acres (2,653 hectares) of federal property 16 mi (~26 km) northwest of downtown Denver, Colorado. The 385-acre (156-hectare) main production area of the RFP, within the controlled area is surrounded by a 6,150 acre (2,491 hectare) buffer zone, which delineates the current RFP boundary.

The State of Colorado is currently conducting a multi-phased study of the possible public health effects of historical operations at the Rocky Flats weapons facility. Phase I of the study, which was funded by the Federal government but under the exclusive control of the Colorado Department of Public Health and Environment (CDPHE), was performed by ChemRisk (a division of McLaren/Hart Environmental Engineering).

During Phase I, an extensive investigation of past operations and releases from the plant was conducted. The Phase I effort produced an assessment of the primary materials of concern, release points and events, quantities released, transport pathways, and preliminary estimates of dose and risk to offsite individuals. The conclusions from Phase I were released in a public summary document (HAP 1993) and in a series of task reports by ChemRisk.

Radiological Assessments Corporation (*RAC*) was awarded the contract to conduct Phase II of the study, which is an in-depth investigation of the potential doses and risks to the public from important historical releases from Rocky Flats. Recommendations for work to be performed in Phase II are outlined in HAP (1993). The complete list of tasks for Phase II follows:

Task 1. Coordination with ChemRisk, to ensure quick and efficient access to the records and individuals contacted by ChemRisk during Phase I of the project.

Task 2. Verification of the radionuclide and chemical release estimates and associated uncertainties that were developed during Phase I of the project.

Task 3. An independent assessment of the risk from past Rocky Flats operations, using state-of-the-art methods to ensure that risks to the public are carefully identified.

Task 4. Evaluation of historical environmental data, which can provide a basis for risk assessment and for reconstruction of releases.

Task 5. Recommendations for additional offsite measurements, using knowledge gained to ensure that new measurements focus on the most important locations and releases.

Task 6. Support for the public involvement efforts.

PURPOSE AND SCOPE OF THIS REPORT

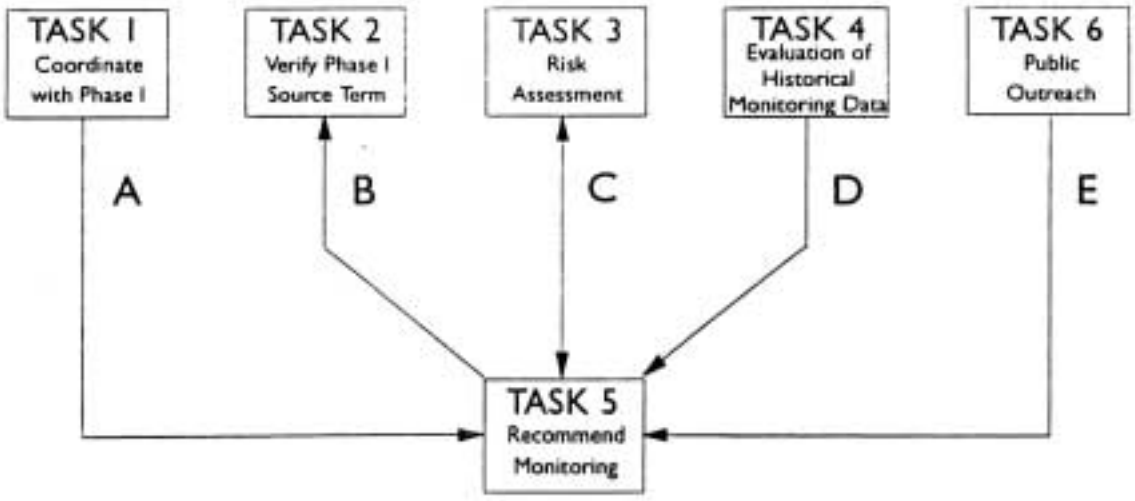
The primary purpose of Task 5 is to recommend new or supplemented studies that could help confirm the results of Phase I and Phase II of the Rocky Flats Plant Historical dose reconstruction. Ideally, the studies identified by this task will provide data than can be used to

1. Verify and /or refine existing source terms
2. Verify some past monitoring results and clarify associated uncertainties
3. Provide evidence regarding past contamination offsite and
4. Provide new data for model calibration or validation.

Monitoring is usually narrowly defined as the use of instruments, systems or special techniques to measure liquid, and/or airborne effluents and contaminants. As such, the term is not strictly appropriate for all types of activities that are recommended in this report. However, in order to be consistent with the title of this task, we loosely apply the term monitoring to include field and laboratory research studies that can be developed to support the dose reconstruction.

It should be emphasized that any monitoring activities conducted as a result of Task 5 are elements of an integrated historical dose reconstruction process. The relationship between Task 5 and the other Tasks in Phase II is shown in Figure 1.

This report presents the results of Part 1 of Task 5. The final Task 5 report will incorporate Parts 1 and 2 and will be submitted before the conclusion of Phase II. Task 5 was divided into two parts to accommodate the review of relevant data not available to the Phase I contractor and to allow adequate input from the Health Advisory Panel and members of the public. The additional data includes previously unexamined archived Rocky Flats documents and the results of current research being conducted by EG&G Rocky Flats and Colorado State University (CSU). Part 1 of Task 5 focuses on identification of data needs and study recommendation, and the selection of the most appropriate recommendation. Part 2 will involve the development of more detailed sampling strategies. This approach is discussed in the section that follows.



Notes on relationships between tasks:

- **A.** Conclusions and recommendations made by the Phase I contractor are considered in formulating monitoring recommendations.
- **B.** Environmental data needed to verify or refine source terms are being identified during this task. If the data are not available in the monitoring records, new or supplemental monitoring is recommended.
- **C.** Environmental data will assist in defining model structure and parameters for risk assessment. Data sets for model validation are being developed. If they are not available from existing documents, recommendation for collecting additional data are made.
- **D.** Data are being evaluated in terms of quality, a completeness, and usefulness as validation data for model calculations, as source term data, or as indicators of spatial or temporal trends. Data deficiencies may indicate the need for additional monitoring.
- **E.** Public recommendations have been provided via interaction with the Citizens Environmental Sampling Committee, public workshops and other means.

Figure 1. Relationships between Task 5 and other tasks in Phase II.

APPROACH

Task 5 is one of the first tasks scheduled to be completed in Phase II of the dose reconstruction. For that reason we cannot fully anticipate specific data needs that may be identified as the work progresses. New data needs and studies proposed after this report is released will be evaluated and addressed in Part 2 and the final report. Thus, the process of selection studies to support the dose reconstruction will remain dynamic, taking account of any new information or insights.

The identification of data needs and the associated development of recommendations for new or subsidiary studies require a multifaceted approach that considers all relevant information. The systematic approach used to conduct Task 5 is illustrated in Figure 2 and is described in the test that follows.

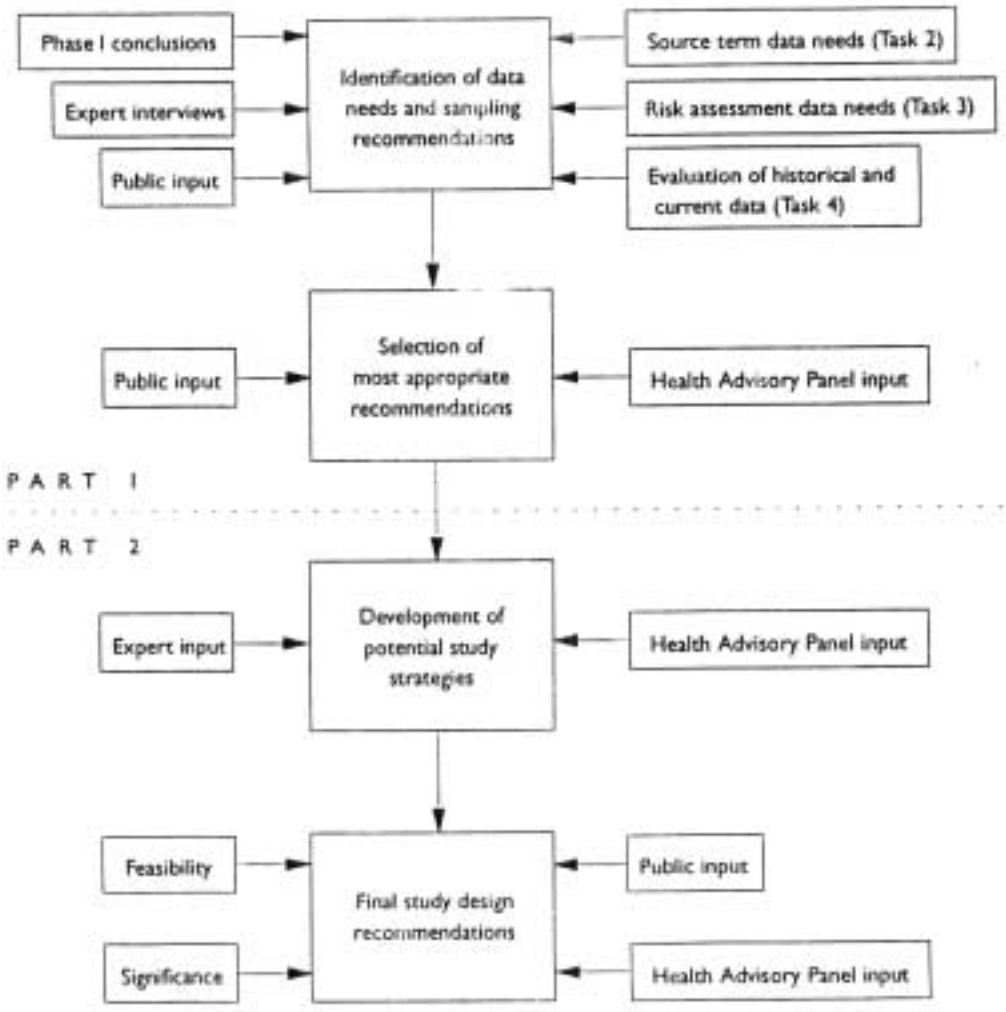


Figure 2. Approach used to conduct Task 5.

During the Part 1 of Task 5 data needs and studies that could be used to support the dose reconstruction were identified from various sources of information. These sources included

1. Phase I documents and discussions with the Phase I contractor;
2. interviews with monitoring experts involved in current and past RFP studies;
3. input from members of the public via the Citizens' Environmental Sampling Committee (CESC) of the Rocky Flats Health Advisory Panel (HAP) and personal communication with select individuals;
4. data reported in historical monitoring documents and collected recently from the Rocky Flats environment by EG&G Rocky Flats, the Colorado Department of Public Health and Environment and Colorado State University, as part of Task 4 of Phase II; and
5. suggestions by *RAC* researchers, based on data needs identified during the conduct of Tasks 2 and 3 (i.e., verification of Phase I source terms and Phase II risk assessment).

The conclusions about Phase I presented by the HAP (1993), were the primary source of data needs and potential studies identified from that portion of the historical dose reconstruction. In addition, Stephen Ripple of ChemRisk, the project manager of Phase I, was queried concerning his thoughts on Task 5.

Monitoring experts who were interviewed for this task are presented in Table 1. These individuals were selected because of their knowledge of and experience in studies involving contaminant transport in the environment around the RFP. They were questioned about their involvement in these studies and their suggestions for studies that would help fill data gaps and expand our knowledge of historical releases and subsequent fate of contaminants in the environment. Other technical experts may be consulted before the release of the final Task 5 report, based on further suggestions by the HAP or the public. For example, Edward Martell of the National Center for Atmospheric Research, has agreed recently to be interviewed by *RAC*. This interview was suggested by some concerned citizens.

Table 1. Individuals Interviewed for Part I of Task 5

Name (affiliation)	Area of expertise	RFP experience
Frank Gifford (Consultant)	Meteorology	Member of the HAP
Michael Guillaume	Environmental engineering	Past manager, Operating Unit 3 (OU3) studies for Remedial investigation
Shawki Ibrahim (CSU)	Radiochemistry	Radioecology studies at RFP; bioassay studies
Phil Krey (DOE Environmental Measurements Laboratory)	Soil science	Numerous soil, air, and sediment studies around RFP
Gerhard Langer (formerly of DOW Chemical Co., currently private consultant)	Resuspension	Resuspension studies at RFP
Iggy Litaor (EG&G Rocky Flats)	Soil science/contaminant distribution	Detailed studies of Pu, Am, and U in onsite and offsite soils for the current Remedial Investigation
Jeb Love (CDPHE)	Surface water hydrology	Rocky Flats Project for CDPHE
Gregory Marsh (Consultant)	Environmental chemistry	Member, Citizens Environmental Sampling Committee
Stephen Ripple (ChemRisk)	Historical dose reconstruction	Phase I project manager
Dave Schoep (CSU)	Sediments	Sediment studies of lakes
Scott Webb (CSU)	Soil sampling	Soil studies downward of 903 Pad area
Ward Whicker (CSU)	Radioecology	Radioecology studies at RFP
James Whiting (EG&G Rocky Flats	Soil science	Background studies around RFP

Public input was sought. First, ideas for sampling studies were obtained from attending meetings of the CESC. Second, solicitation of public input was made during a presentation of the draft Task 5 Report to the HAP in December of 1993. The draft version of this report was also distributed for comment to individual members of the CESC and was made available to other members of the public during public workshops. In addition, personal contracts were made with the Environmental Information Network concerning data needs and ideas for further studies.

Historical and current monitoring documents reviewed for Task 4 are maintained in a database, which is available for public review. An updated list of titles is provided in the briefing book for the November 1994 meeting of the Health Advisory Panel. Some of the documents that ate especially relevant to Task 5 are presented in the References section at the end of this report. For Task 5, particular attention was focused on studies that attempted to evaluate the offsite impact of RFP. These studies provided much of the theoretical basis for past estimates of source terms and for current understanding of the fate and transport of the contaminants released to the Rocky Flats environment. During the document review, *RAC* researchers attempted to identify data gaps and anomalies, particularly in light of other Phase II tasks.

Task 2 and 3 activities are focused on the following source terms: (1) plutonium released during the 1957 fire, (2) plutonium released from the 1969 fire, (3) routine plutonium releases, (4) historical releases of plutonium from the 903 Pad area, (5) carbon tetrachloride, (6) beryllium, and (7) plutonium and tritium in surface water. *RAC* researchers have considered data needed to verify or refine these source terms and to calibrate or validate source release and transport models.

The second element of our approach was to assess monitoring recommendations for limitations in their practical application or usefulness to the dose reconstruction. The general process used to accept a study is illustrated in Figure 3.

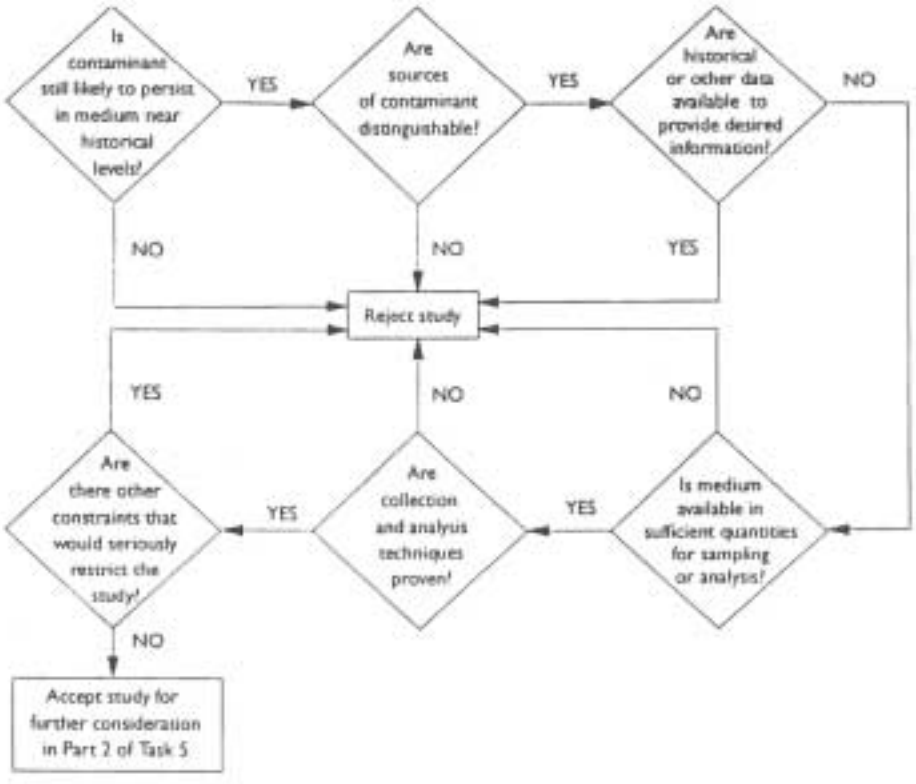


Figure 3. General process used to select or reject a proposed study.

The final stage of our approach (Part 2 of Task 5) will be to develop possible implementation strategies for each of the studies recommended in Part 1. We will then select the study designs that are most feasible, in terms of time, cost, and other factors, and that will provide the optimum data sets for supporting credible dose calculations. The final phase of Task 5 will also provide additional opportunities for input from monitoring experts and the HAP.

IDENTIFICATION OF DATA NEEDS AND POTENTIAL STUDIES.

This section presents a discussion of how specific data needs and potential studies were identified. The discussion is presented in the following topical order: (1) Directions from Phase I, (2) Recommendations from Monitoring Experts, (3) Areas of Public Concern, (4) Conclusions from Document Review (Task 4), and (5) Data Needs Identified During the Conduct of Tasks 2 and 3.

Directions from Phase I

Phase I conclusions which could potentially impact Task 5 of the Phase II study (HAP 1993) are as follows:

1. Twelve chemicals and radionuclides were found to be major contributors to offsite exposures. They are: beryllium; carbon tetrachloride; chloroform; methylene chloride; tetrachloroethylene; 1,1,1-trichloroethane; trichloroethylene; americium, plutonium, thorium, tritium and uranium. Of these, five contaminants (americium-241, beryllium, thorium-232, isotopes of plutonium, and isotopes of uranium) are most likely to persist in measurable amounts in the environment.

2. The events associated with the most significant contaminant releases from Rocky Flats are: plutonium releases from the 903 Pad area; plutonium releases from the 1597 fire; and routine releases of carbon tetrachloride.

3. Although routine releases of plutonium and plutonium releases from other non-routine events have been apparently minor (compared to those from the 903 Pad and the 1957 fire) reasonable questions have been raised about the quality of routine monitoring programs and about the potential for unmonitored recurrent releases.

4. Data available to date indicate that contaminants in groundwater have not yet moved offsite. [Note: Results of groundwater monitoring in the Indiana Street boundary wells during 1992, the year that the plant suspended operations, show that volatile organic compounds (VOCs) and dissolved metals were detected in concentrations slightly above detection limits in alluvial, or shallow wells. Radionuclides detected in these boundary wells were attributed to high suspended solids derived from stream sediments (EG&G 1992). There is no direct hydraulic connection between the shallow alluvial aquifer and deeper aquifers used for domestic water supplies.

For the period under consideration for dose reconstruction activities (1953 through 1992), no offsite contamination of groundwater is evident.]

5. Surface water appears to have been an intermittent and apparently minor source of offsite contamination from Rocky Flats. However, because of limitations in available data and in information on process waste streams, this pathway is being evaluated in Phase II.

6. Phase I did not perform an extensive analysis of potential exposures to sediments in Great Western Reservoir or Standley Lake. Continuing public concern for plutonium in lake sediments indicates that additional evaluation may be warranted.

Phase I also identified the following areas for additional study in Phase II that involve Task 5:

1. An independent assessment of releases from the 1957 fire

2. An independent assessment of releases from the 903 Pad area

3. Further evaluation of the 1969 fire

4. Further evaluation of the quality of historical monitoring data

5. Further assessment of releases to surface water

6. Further evaluation of releases to lake sediments

7. Further evaluation of potential exposure and health risks of beryllium

8. Continuing evaluation of resuspension of deposited plutonium and other contaminants. (Note: Phase I identified resuspension of deposited plutonium as a key uncertainty in the use of environmental monitoring data, because estimated resuspension factors may vary by many orders of magnitude.)

The conclusions and recommendations resulting from the Phase I study imply that any further monitoring should focus primarily on plutonium, beryllium, and carbon tetrachloride. Furthermore, studies should focus on environmental media, which could best provide historical records of releases via air and surface water [i.e., air (archived filters), soil sediment, or long-lived vegetation.] Groundwater is specifically excluded, as there is no evidence of contaminant transport offsite during the period of concern for dose reconstruction. Finally, studies to better define key transport processes such as resuspension from Rocky Flats soils may be appropriate.

Recommendation from Monitoring Experts

Discussions with monitoring experts provided many potential study suggestions. These suggestions, along with the individual responsible for the suggestion, are presented in Table 2. Note that some of the individuals interviewed (as listed in Table 1) either did not contribute suggestions or shared ideas that were proprietary in nature and could not be reported here.

Table 2. Studies Recommended by Monitoring Experts

Name (Affiliation)	Suggestion	Justification
Shawki Ibrahim (CSU)	Continue with urine bioassay program. Fifty samples have been collected and half have been analyzed for ^{239}Pu. CSU recommends doubling the sample size.	Program in place; well accepted by the public; useful for determining total body burden.
	Collect and analyze unclaimed ashes from crematoria for Pu.	Can yield total body burdens for many periods of time. CSU has pre-RFP samples.
	Analyze more soil samples (current and archived) for $239_{Pu}/240_{Pu}$ ratios to establish background. Alternatively, collect more samples from remote locations and assess variability of fallout Pu.	Few studies (HASL, CSU) have distinguished RFP Pu from fallout Pu using isotopic methods. ^{239,240}Pu concentrations from fallout can vary in this region of the US from 0.001 to 0.08 pCi/g, depending on regional and local weather patterns, particle sizes of Pu and varying erosion and transport conditions. Thus, the full extent of RFP contamination is still unknown. Archived RFP soils may still exist. (Note: RAC determined that CSU, Krey, and Illsley soil samples are archived.)
	Analyze lichens in the RFP environment, particularly in the 903 Pad area for Pu.	Lichens are long-lived and may provide a spatial record of deposition and resuspension of Pu. CSU recently completed a study in the vicinity of the 903 pad, which evaluated Pu in lichens as function of distance and direction from the 903 Pad. Spatial distribution was similar to that found in surface soils. Lichen analysis may be useful for verification of soil study data.
Gerhard Langer (previously DOW Chemical Co., currently private consultant)	Conduct “fresh” resuspension studies involving ZnS to derive model of resuspension from 903 Pad during grading activities in 1969. Studies would involve lab and field work with bare soil and sod.	Most of the release from 903 Pad area occurred during the grading activities. Over time significant resuspension, even at high winds, could not be measured most likely due to the soil becoming crusty over time, and to protection of soils by the grass canopy.
	Conduct isotopic analyses of Pu in soil to distinguish RFP Pu from fallout Pu.	Fate of RFP Pu can be implied from results.

Table 2. Studies Recommended by Monitoring Experts

Name (Affiliation)	Suggestion	Justification
Jeb Love (CDPHE)	Develop a comprehensive program to collect and date sediment core layers, determine $^{239}Pu/^{240}Pu$ ratios, and study physical characteristics of associated particles. Include all reservoirs near the RFP (Great Western, Mower, Marshall, and Standley). Establish temporal and spatial patterns.	Current EG&G studies are being conducted to comply with (Resource Conservation and Recovery Act) RCRA requirements and may not be compatible with dose reconstruction needs. Past Health and Safety Laboratory (HASL) studies showed definite temporal patterns, which could be correlated with RFP events. Sedimentation rates were derived as well as Pu contribution from surface runoff from RFP. New studies could be used to deduce the origin (including surface water runoff), mechanisms, and transformations that occurred over time.
Scott Webb (CSU)	A study of Pu concentrations south of the 903 Pad area towards Woman Creek to determine if Pu has been transported via surface erosion and deposited in that area.	A re-evaluation of Pu in soil plots studied by Little (1976) 15 years earlier show a decrease of 25% in Pu concentration in the surface layer and no difference at lower depths. One theory is that the loss is due to surface erosion.
	An *in situ* simulated rainfall and lab study of downward leaching of Pu would help elucidate downward transport theory.	Initial deposition and rapid, downward transport may explain why layers of soil below the surface layer did not appear to differ from the previous study conducted by Little (1976).
	Analyze samples by mass spectrometry to distinguish ^{239}Pu and ^{240}Pu and thus obtain isotopic ratios in 903 Pad area.	Isotopic ratios can be used to distinguish releases from 903 Pad area and global fallout.
Ward Whicker (CSU)	Include a profile of ^{137}Ca in any soil studies to help interpret Pu data.	Can be used to indicate whether or not soil has been disturbed.
	More Pu measurements need to be made in 903 Pad area, inside the buffer zone fence, and compared to past measurements - paying special attention to slope and interceptor ditch at bottom. Also, need to look at Pu isotopic ratios with distance.	Decrease in Pu observed between the studies of Little et al. (1980) and Webb (1992) could be explained by surface water erosion. Isotopic ratios will help distinguish Pad 903 area and global fallout Pu.
	Analyze archived air filters from Station S-8, which is downwind from 903 Pad, for Pu	The results can be used to calculate a time-integrated concentration of Pu in air and with soil results to derive a deposition velocity.
	Analyze archived air filters and soil from other air monitoring stations for Pu.	May be possible to estimate the resuspension rate and therefore air concentrations and dose.
	Conduct site-specific percolation studies.	Critical parameter for determining the availability of Pu for surface soil transport.

Areas of Public Concern

The greatest area of public concern, based on opinions expressed at CESC meetings, is that independent sampling and analysis of soil be conducted to (1) compare results using sample areas of private concern. The CESC has been specifically tasked to design and implement a soil sampling program to achieve these goals. As such, it would be redundant to include new soil sampling to address public concerns as part of the Task 5 recommendations.

One area of interest expressed by some CESC members was the use of vegetation to provide a temporal record of releases from the RFP. A recent article (Kudo et al. 1993) demonstrates that tree rings can provide a record, albeit qualitative, of discrete events involving the release of plutonium. In this study, tree core samples collected near the 1945 nuclear detonation site of Nagasaki, Japan showed elevated plutonium concentrations in the years corresponding to that detonation, as well as the subsequent global fallout events.

There is one grouping of old apple trees on the RFP that are known to predate the plant and offer some quantity for a similar study. However, these are located approximately 1 mile SW of the 903 Pad area and are not considered to be downwind of that area (plutonium concentrations in soil indicate that deposition occurred in a direct easterly path). The grove may have intercepted plumes from other episodic events, such as the 1957 fire, but this is unknown. As such, we cannot anticipate that levels of plutonium in the tree rings will be above the detection limits associated with conventional radiochemical techniques. The most sensitive method for plutonium detection at this time is the track etch method. This method has been used successfully on fluids, as evidenced by the CSU urine bioassay study. However, it has not been developed for woody material. It is not known if a satisfactory transfer of this analytical technique can be achieved. The development costs, combined with the standard laboratory analytical cost of $1500 per sample, would probably be prohibitive for the quantity and quality of data attained.

Other trees do exist on the site. Cottonwood trees follow both Woman and Walnut Creeks northeast and southeast of the 903 Pad area. Those located south and southeast of the 903 Pad area are the closest to the site, about 0.25 miles distant. Most of these trees appear to be too young for our purposes. In the easterly direction, there are some Siberian Elms along Indiana Street (about 0.5 miles east of the 903 Pad area). However, there appear to be few of sufficient age, making a good statistical design difficult to develop.

The use of trees as a temporal record of plutonium releases is probably a good research project, however, of limited value to the dose reconstruction. The analysis of sediment cores for plutonium and radionuclides (such as Cs-137 and Pb-210) that help to date layers is a preferred, proven method of determining plutonium releases over time.

Conclusions from Review of Historical Monitoring Documents (Task 4)

Historically, environmental monitoring and research studies have focused on plutonium in air, soil, surface water, and sediment. Because soil and sediment act as “sinks” (accumulators) for long-lived contaminants, our interest for future monitoring recommendations lies primarily with those media. A brief summary of research conducted to date and specific conclusions, which were considered in formulating additional monitoring needs, are presented below, by media. Note that conclusions and recommendations identified during Phase I, which were listed previously, are not repeated here.

Soil

Soil represents a significant sink for plutonium and other long-lived radionuclides and metals. Little et al. (1980) estimated that the soil contains 99.7% of the plutonium in the grassland ecosystem at Rocky Flats. Numerous soil studies have been conducted and are being conducted at and around the RFP. A number of these were reviewed in the December 1993 Task 4 Progress Report (Rope et al. 1993). Table 3 summarizes some of the major soil research studies, listed in order of occurrence, conducted to date at or near the RFP. This list is not intended to be inclusive of all soil sampling programs conducted at or near the site, but represents the historical studies most often cited in summary documents and used for source term estimates and risk assessments. Recent and ongoing studies that should be examined prior to making final monitoring recommendations are also included. Further work on Task 4 may involve additional reviews of soil studies.

From reviews of historical and recent studies of soils around the RFP, we have identified a number of potential data needs for the Project that we address here. These potential data needs are (1) concentrations in soil of contaminants other than plutonium and americium, especially beryllium, (2) precise determinations of background concentrations of contaminants in the soil at the RFP, (3) quality assurance and other data to allow the comparability of the various studies, which were completed at different times using different methods, (4) information on the total inventory and spatial distribution of Pu in soils, from RFP releases, (5) information on the fate and transport of actinide contaminants in the soil column, and (6) Pu concentrations in soil around the RFP for the earlier years of site operation.

One of the obvious conclusions from reviews of the historical soil studies is that almost all of the studies focused on measurements of radionuclides in soil, particularly plutonium and americium. However, we are also concerned about releases of and subsequent exposure to people by other contaminants. Soil may be a sink for some of these contaminants, such as beryllium. Since the available monitoring for beryllium is fairly limited, one potential data need is additional monitoring of beryllium in soils. Such information could be used to help develop or verify the beryllium source term. In deciding whether a study of beryllium in soils should be proposed, two criteria argue against such a study at this time. A preliminary review of the limited beryllium monitoring data, from our draft Task 4 report (Rope et al. 1993) indicated that onsite concentrations of beryllium may not be greater than off site concentrations. Second, monitoring of beryllium in stack effluents has been routinely performed in the past (Werkema 1980), and these data may be sufficient for reconstructing the beryllium source term. Thus, a study of beryllium concentrations in soil is not recommended.

A number of the historical soil studies included investigations of background concentrations of plutonium (generally ^{239,240}Pu). We reviewed many of the previous studies on background concentrations in a Technical Memorandum (Schmidt 1994). Based on that review, it was concluded that the existing data were sufficient for a rough estimate of the background concentration, to be used for general perspective. Thus, one data need is the clear definition of ambient levels of plutonium around the RFP. A more precise determination of the background plutonium concentration would allow a better calculation of the extent of plutonium contamination that is due to RFP releases. The isotopic studies (using ratios of ^{239}Pu to ^{240}Pu, and other ratios) conducted by HASL (Krey and Krajewski 1972a; Krey 1976) demonstrate that the use of mass spectrometry is one potentially viable way to distinguish plutonium released from

the RFP and from fallout. A new study could examine more samples (new and archived) using this or similar techniques, to determine the plutonium background around the RFP with greater certainty.

Table 3. Summary of Some Important Soil Sampling Studies Reviewed

Agency (reference)	Number and location of samples	Depth of samples	Analyses	Comments
RFP (Thackeray 1953, Hammond 1957, Hammond 1958, and others	Onsite and off site locations		gross alpha	Results are less useful than from other studies, because no Pu results were obtained
CCEI and NCAR (CCEI 1970, Poet and Martell, 1972)	18, within 6 miles of RFP	1 cm	^{239,240}Pu, ^{90}Sr	Presented first public proof of off site contamination.
HASL (Krey & Hardy 1970)	33, up to 40 miles downwind of RFP	To 20 cm	^{239,240}Pu	Study used most often in past source term estimates.
HASL (Krey and Krajewski 1972a)	2. about 10 and 20 miles from RFP	10 cm	^{238}Pu,^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu	Demonstrated feasibility of using isotopic ratios to separate RFP and global fallout concentrations
CSU (Little 1976)	10, from two plots near 903 Pad	21 cm	^{238} Pu, ^{239,240}Pu	Pu concentration s and soil particle size ranges measured in 3-cm segments of each deep sample.
CSU (Little and Whicker 1978)	15, transects near plots of Little (1976)	21 cm	Plutonium	Pu concentrations and soil particle size ranges measured in 3-cm segments of each deep sample.
HASL (Krey et al. 1976a)	26, east of RFP to Indiana Street	10 cm	$^{239, 240}$Pu, ^{241}Am	Soil Pu concentrations related to air sampler
HASL (Krey 1976)	19, downwind of RFP to Denver	10 cm	^{239}Pu, ^{240}Pu	Able to identify source of Pu (RFP vs. Fallout), using isotopic rations.
HASL (Krey et al. 1976b)	27, up to 3 km downwind of 903 Pad	20 cm	^{239,240}Pu, ^{241}Am	Pu and Am studied with depth.
CDPHE (CDH 1977)	13, outside RFP to a distance of 100 miles	To 0.32 cm	^{137}Ca, ^{239,240}Pu	Includes comparative evaluation of "Dr. Johnson sweep" method with CDH methods.
CDPHE (CDH 1990)	13 near and 8 remote for most years	0.48 cm	^{239,240}Pu	Summary of 1970-1989 results
CSU (Webb 1992)	10, downwind of 903 Pad	To 21 cm	^{238}Pu, ^{239,240}Pu	Repeated earlier study of Little (1976) to determine changes.
EG&G (Litaor 1993, DOE 1993)	118 surface plots and 26 pits within 3 km from the RFP	To 64cm and 96 cm (for pits)	^{239,240}Pu, ^{241}Am, U	Geostatistical analysis of surface data. Test pits to investigate vertical distribution and transport processes of actinides.
EG&G (Whiting 1994)	50	5 cm		Study in progress. To evaluate background concentrations.
CDPHE (Love 1994)	13, within 7 miles from RFP	To 0.64 cm	^{239,240}Pu	Summary of results for 1970-1991.
CDPHE (Jones et al. 1994)	13 within 7 miles, and 9 remote	To 0.64 cm	^{239,240}Pu	Summary of results for 1970-1991,, with statistical comparison of data sets and contour plots.
CSU (Whicker 1994)	on site, off site and background locations	0.3 cm 3 cm, and to 21 cm	^{239,140}Pu, ^{241}Am	Study in progress. Includes depth profile samples, and background samples along the Front Range.

Alternatively, a less expensive approach would be to establish a study focused strictly on sampling remote locations and assessing the variability of fallout plutonium concentrations. In fact, there are two current studies focused on obtaining a clearer picture of background

plutonium levels. A project by CSU includes the measurements of soil plutonium concentrations at ten locations along the Front Range of the Rocky Mountains, from north of Fort Collins, south to Colorado Springs (Whicker 1994). Results of this study are expected soon. Also, a study by the RFP of background concentrations involves sampling at 50 locations (Whiting 1994). A report of results for this study is expected in early spring 1995. These two current studies should add considerably to the knowledge about background concentrations of plutonium in soils. Thus, additional sampling for background determinations is not warranted.

Recent environmental monitoring studies have generally employed and documented extensive quality control (QC) and quality assurance (QA) measures. For some of the historical studies, however, QA and QC information may not be documented, and in some cases may not have been performed. In addition, sampling and analysis methods have changed over the years. Thus, standard concerns with the use of data from historical studies are the over all quality of the data, and comparability between studies. To have more confidence in the historical studies, additional QA and QC data would be useful. This could be accomplished by re-analyzing archived soil samples from the historical studies.

As discussed elsewhere in this section and indicated in Table 3, there are large amounts of data being generated in the recent and current sampling programs, particularly those of the RFP and of CSU. These newer studies are utilizing appropriate QA and QC protocols, and will provide data useful to various aspects of our work. The additional data will, to some extent, reduce reliance on the historical studies. Jones et al. (1994) analyzed data from the CDH soil sampling of 1970-1991, along with data from RFP soil monitoring from 1984-1992. Results of their statistical analyses indicated no significant trends in plutonium concentrations with time. While these analyses did not encompass all soil sampling performed around the RFP, their results indicate that the plutonium concentrations in soil have probably not changed appreciably since the early 1970s. Thus, it is possible to compare historical data with current data. In addition, the current project by the RFP to investigate background concentrations will also include a comparison of two different analysis methods for plutonium in soil (Whiting 1994). The two general methods are acid leaching of metals from the soil for subsequent analysis, which was apparently used in many of the historical studies, and complete dissolution of the sample (using hydrofluoric acid), which ensures all the plutonium is analyzed. Information from this comparison will also aid in comparisons of the various historical and more recent studies. Because recent and new data will improve our ability to compare results from different studies and reduce our reliance on the historical studies, at this time we do not recommend reanalysis of archived samples to improve confidence in the historical results.

Another data need is a complete description of the plutonium contamination around the RFP, including total inventory and spatial distribution of plutonium in soils. This information may be useful for development or verification of the plutonium source term, and for calibrations or comparisons with predicted (modeled) concentrations. A number of historical studies attempted to assess the total inventory and spatial distribution of plutonium in soils due to releases from the RFP (Krey and Hardy 1970; Seed et al. 1971; Krey et al. 1976b; Krey 1976). More recently, statistical methods have been applied to better determine inventory and spatial distribution of plutonium. For the remedial investigation and feasibility study for potential cleanup of contaminated soils around the site, the RFP has performed new sampling, and has used geostatistical techniques in its data analyses (Litaor 1993; DOE 1993). Sampling for the remedial investigation is continuing. Historical results of the CDH sampling have also been

analyzed using geostatistical techniques (Jones et al. 1994). In addition, the independent Citizen's Environmental Sampling Committee is obtaining and analyzing additional soil samples from around the RFP. Our reviews of the existing studies have not been completed, and additional results are anticipated shortly, from the current studies. However, it appears that results of the existing studies and current studies should be adequate for evaluating plutonium inventory and spatial distribution in soils. Thus, no additional studies for this purpose are recommended.

For environmental transport modeling purposes, another data need is information related to the fate and transport of plutonium (and americium) in the soil column. This data would be useful for the development of environmental transport models for the exposure assessment. Much research has been performed in this area. Studies of plutonium in soil profiles and in air by HASL (Krey et al. 1976b) (Volchok et al. 1977) and the EPA (1977) indicate decreasing availability of plutonium in surface soils for resuspension, and thus lower airborne concentrations of plutonium, with time. This has been attributed by several authors to stabilization of plutonium in the surface soil. Thus, it is possible that much of the RFP plutonium may still remain in the soil, although it may have been transported further down in the soil column.

CSU researchers theorize, based on studies of plutonium particle size distribution and soil profiles of plutonium near the 903 Pad that the following scenario occurred (Little and Whicker 1978): (1) the plutonium contaminant was in the form of an oxide, (2) the plutonium oxide became attached to soil particles, (3) gusty winds combined with periods of surface disturbance hetereogeneously redistributed the particles to the east and southeast of the Pad, and (4) the soil-plutonium particles were eventually broken down by weathering and were dispersed laterally and downward in the soil profile. A comparison of plutonium concentrations measured in the same plot in 1974 and 1989 indicates similar concentrations at depths below 3 cm (Webb 1992). However, the concentration measured in the top 3 cm in 1989 was 25% of that measured in 1974. Erosion by wind and water was speculated as the main cause for the difference, although differences in analytical techniques and statistical treatment could also contribute to the difference. Webb (1992) suggests that isotopic analyses of soils be conducted down slope of the Pad, to include the runoff interceptor ditch below it.

McDowell and Whicker (1978) hypothesized that, based on a study of plutonium particle frequencies and sizes near the 903 Pad, that the heterogeneity in the spatial distribution of plutonium particles in the soil may partially explain observed plutonium concentrations in the area. A study conducted in 1975 by Hayden et al. (1994) on particle sizes of plutonium in the RFP buffer zone east of the plant indicates that larger particle sizes are associated with the 903 Pad and smaller particle sizes (mean diameter of 0.08 micron) with stack releases. These two studies confirm the usefulness of measuring particle sizes and isotopic ratios in RFP soils to help elucidate the fate of plutonium in soils.

The current remedial investigation studies by the EG&G Rocky Flats will provide additional information on plutonium transport pathways in soil. Some information from this study, that will be helpful for determinations of inventory and spatial distribution of plutonium in soils, was discussed elsewhere (Litaor 1993). In addition, results of investigations utilizing 1 m deep test pits may be helpful toward understanding the fate and transport of plutonium (and other radionuclides) in the soil column. In this study, 26 pits were dug and investigated, mostly northeast to southeast from the 903 barrel storage pad (DOE 1993). The vertical distributions of

^{239,240}Pu and ^{241}Am were evaluated. Also investigated were mechanisms of vertical and lateral redistribution of plutonium and americium in the soil, as well as physical and chemical parameters that may be important to understanding transport in soil. Results of this study indicate that 90% of plutonium is in the upper 12 cm of the soil column; however, transport to greater depths than previously reported at the RFP was observed at in at least one pit. It was also concluded that biological activities, such as those of earthworms and burrowing animals, were a significant fact or in the transport of plutonium and americium in the soil column.

The results of approximately 60 more soil samples (west of Indiana Avenue) and 38 soil pits will greatly enhance our knowledge of the fate and transport of plutonium, americium, and uranium in soils. The pit studies include measurement of soil characteristics, the use of a Soil Water Monitoring System (SWMS) to measure actinide migration during natural and simulated rainfall events, and the examination of pedogenic processes on vertical distribution of plutonium and americium.

The early soil monitoring studies around the RFP, before 1969, did not include plutonium-specific analyses; rather, gross measurements of alpha-emitting radionuclides ("gross alpha") were made. From preliminary reviews of these early studies (for example, Thackeray 1953, Hammond 1957, and Hammond 1958), gross alpha concentrations are not greater in on site soil than in off site soil. Thus, these results will probably not be useful for dose reconstruction purposes. A data need is measurements of plutonium (and other radionuclide) concentrations for times before 1969. Archived soil samples, if any exist, could be used for theses measurements. Such data could be useful for development or verification of the source terms associated with the earlier releases, such as from the 1957 and 1969 fires. We have searched for archived soil samples from this time period, but have so far been unsuccessful in locating any. However, if archived soil samples can be found, consideration should be given to having radionuclide analyses performed. Soil samples from both before and after the episodic release events (1957 and 1969 fires) may be particularly useful.

Based on the discussions above, we conclude that the existing soil studies, along with current studies, should generally be adequate for our dose reconstruction purposes. However, we do recommend further consideration of the analyses of archived soil samples from 1969 and earlier, conditioned on the existence of any such archived samples.

Sediment

Because sediment is an effective sink, it contains the major inventory of contaminants released to or deposited on a body of water. Contaminants such as plutonium become associated with suspended particulates in the water column and settle with the suspended sediments at a fairly constant rate. The result is an historical record of pollutant releases in the layers of sediment at the bottom of the impoundment. Data obtained from the analysis of sediment samples can thus provide an integrated measurement of contamination from liquid and/or airborne effluents and an indication of temporal trends, which may be used to identify specific release events.

Many studies have been conducted to determine plutonium and heavy metal concentrations in lakes, ponds, and creeks around the RFP. Some important studies are summarized in Table 4. Generally, routine sediment monitoring studies did not provide data useful to the dose reconstruction. Few samples were collected, locations and methods were often not identified, and

results were not always reported. Thus, the routine monitoring reports are not included in Table 4.

The studies summarized in Table 4 demonstrate that lake sediments show definite temporal and spatial patterns of plutonium concentrations, which indicate historical trends. The use of isotopic ratios of specific radionuclides and specific dating techniques (e.g., the use of ^{137}Cs and ^{210}Pb) have provided fairly accurate information on the sources of plutonium (i.e., liquid effluents, deposition of airborne releases, or fallout) and dates of deposition. It is apparent that sediment can provide data that are very useful to the dose reconstruction. Some potential data needs identified during the document review include (1) concentrations in sediment of contaminants other than plutonium and americium, especially beryllium, (2) concentrations of plutonium isotopes and other contaminants of concern in sediment profiles of downwind and downstream lakes, and (3) adequate background data.

The December 1993 Task 4 Progress Report (Rope et al. 1993) notes the potential need for the measurement of beryllium and carbon tetrachloride in sediment cores. Historical monitoring data are very limited for these two contaminants. There is a small possibility that carbon tetrachloride may have persisted in very anaerobic and rapidly deposited sediments. However, it is highly unlikely. For this reason we do not recommend analyzing sediment samples for carbon tetrachloride.

Sediment would be a sink for beryllium and could be analyzed for that contaminant to quantify the depositional history. However, there are two major arguments against conducting such a study. First, there are multiple airborne sources of beryllium in the region, which would make it impossible to distinguish the source of the beryllium measured in the sediment. Second, beryllium concentrations measured recently in the sediments of Great Western Reservoir (the only impoundment which historically received liquid effluents from the RFP) by EG&G Rocky Flats (EG&G 1994) are within the range of background concentrations. In addition, EG&G Rocky Flats found no discernible spatial patterns in the sediment, indicating a natural population. These findings imply that there is no evidence of significant concentrations of beryllium being released from the RFP to the Great Western Reservoir via air or water. Therefore, a study of beryllium in sediments is not recommended.

Table 4. Summary of Some Important Sediment Sampling Studies Reviewed

Organization/reference	Location(s)[a]	Sampling method	# of samples	Nuclides
NCAR/Poet & Martell (1972)	GWR, CL, Boulder R., Boyd Lake, HP	Not reported	8	Pu, ^{241}Am (GWR & HP)
EPA/EPA (1971)	GWR, SL, MR, CL, WLC, WMC	Hand trowel	16	Pu, ^{90}Sr, U, Gross a, Total a radium
EPA/EPA (1973)	MR, CL, WLC, WMC	Hand trowel	9	Pu
	GWR, SL	Dredge	22 (20-GWR)	Pu
	GWR, SL	Core	14 (12-GWR)	Pu
EPA/EPA (1975)	GWR, SL, RR, Marston Lake	Dredge	46 (20-GWR, 17-SL)	^{238}Pu, ^{239}Pu[b], ^{137}Cs, ^{89}Sr & ^{90}Sr(13)
	Cherry Creek R.			
	GWR, SL	Core	24(15-GWR)	^{238}Pu, ^{239}Pu
DOW/Kunert & Werkema (1974)	GWR, SL	Dredge, Core	-1/3 of EPA (1975) Samples (15)	^{239}Pu, ^{137}Cs
CSU/Johnson et al. (1974)	HP	Polyvial (composites of 5 gm samples collected monthly)	17 to 24 at each of 7 HPs (-1/2 before and -1/2 after pond reconstruction)	^{239}Pu
	WLC	Dredge	8	^{239}Pu
	HP	Core	18	^{239}Pu
PNL/Thomas & Robertson (1981)	GWR, SL	Core	9	^{239}Pu, ^{137}Cs
EML/Hardy and Volchok (1980)	SL	Core	1	^{238}Pu, ^{239}Pu, ^{137}Cs, ^{241}Am
Rockwell/Hurley (1979)	GWR spillway	Hand trowel	14	^{239}Pu, ^{241}Am
		Core	14	^{239}Pu, ^{241}Am
Rockwell/Hurley (1980)	GWR spillway	Core	7	^{239}Pu, ^{241}Am
Rockwell/Rockwell (1985)	GWR	Dredge	43	^{239}Pu
		Core	4	^{239}Pu
Rockwell/Rockwell (1984)	SL	Dredge	63	^{239}Pu
CO School of Mines/Cohen et al. (1990)	Wellington Lake, Halligan Res.	Core	2	^{239}Pu, ^{137}Cs, ^{210}Pb
CSU/Whicker (1994)	GWR	Core	50–60	^{239}Pu, ^{137}Cs
EG&G Rocky Flats/EG&G (1994)	GWR, SL, MR (Note: some samples collected at Rockwell sample sites)	Dredge	94	Metals and radionuclides including be, ^{137}Cs and isotopes of Pu, Am, and U
	GWR	Core	46	Same as above

[a]GWR = Great Western Reservoir; SL = Standley Lake; MR = Mower Reservoir; CL = Calkins Lake; RR = Ralston Reservoir; HP = RFP holding ponds; WLC = Walnut Creek; WMC = Woman Creek

[b]Unless specified otherwise, ^{239}Pu is actually ^{239}Pu plus ^{240}Pu.

The majority of data on plutonium have been collected from the Great Western Reservoir. Initial studies in the 1970s (EPA 1971, 1973, 1975; Kunert and Werkema 1974; and Thomas and Robertson 1981) indicate dramatic changes in the surface concentration patterns and an increase in total plutonium inventory. These changes can be attributed to reconstruction of the B-series holding ponds at the RFP, between 1970 and 1973, which caused an influx of sediment resuspended from the holding ponds into the Great Western Reservoir. These ponds, which received process waste in the early years, contain significant inventories of plutonium (Johnson et al. 1974). Since late 1974, the intended practice has been to keep all process wastewater on the Plant site where it is evaporated. Thus liquid effluents have not impacted Great Western Reservoir sediments as significantly since that time. Studies performed after 1974 support this conclusion as plutonium concentrations in surface sediments decreased dramatically in the 1980s (Thomas and Robertson 1981; Rockwell 1984; EG&G 1994; and Whicker 1994).

Core samples collected by various researchers show that airborne deposition also added to plutonium concentrations in sediment of the Great Western Reservoir. Thomas and Robertson (1981) showed depth distributions of ^{239}Pu and ^{137}Cs, which clearly indicate a large increase in 1969, possible, correlated with the 903 Pad area release. Similar patterns were derived from data reported in Rockwell (1984), although core sections could not be dated due to the lack of markers, such as ^{137}Cs or ^{210}Pb measurements.

It would be useful to collect additional cores from Great Western Reservoir and accurately assess the depositional history of plutonium, by dating the cores with Pb-210 and Cs-137 and analyzing cores for specific isotopes of plutonium. However, Colorado State University (Whicker 1994) recently collected numerous cores from Great Western Reservoir in 1993 and found only a few cores with any discernible temporal patterns in plutonium concentrations. There is no explanation for why the sediment layers in these samples are indistinct. The sediment may have been mixed. The current effort by EG&G Rocky Flats to assess Great Western Reservoir sediments may provide additional information. We are not recommending any new sediment sampling at Great Western Reservoir at this time for two reasons. First, the CSU results indicate that the sediment layers are no longer intact and will not add to the information we already have from past studies. Second, additional data are being obtained by EG&G Rocky Flats.

Fewer studies have been conducted on Standley Lake, which receives no liquid effluents from RFP. However, the most detailed core study was conducted by HASL (Hardy and Volchok 1980) at Standley Lake. This study demonstrated that isotopic analyses of sediment layers (2-cm core segments were used) can yield information on historical deposition of RFP plutonium and on deposition rates. Discrete events, such as releases from the 903 Pad, are quite apparent. When combined with soil isopleths, deposition from surface erosion and from airborne deposition can be estimated. Unfortunately, the HASL results are based on only one 50-cm core.

Cores collected during other studies on Standley Lake, and Great Western Reservoir, were not as finely subdivided as the one examined by HASL, were not analyzed for specific isotopes ^{239}Pu and ^{240}Pu, and, with few exceptions, were not dated. The need to collect additional sediment cores from Standley Lake and to perform isotopic analyses to ascertain temporal patterns of the actinides is evident. EG&G Rocky Flats has recently collected core samples from Standley Lake and is in the process of analyzing them for specific isotopes of plutonium and uranium, ^{137}Cs, ^{241}Am, and various metals. They also intend to date the cores. However, there is some question as to whether the information will be adequate for use in the dose reconstruction.

The objectives of the remedial investigation is to determine if the sediments currently pose a health hazard to the public. Our objective is to accurately determine historical airborne deposition patterns. Thus, the EG&G Rocky Flats study design may not be compatible with our objective. For this reason, we are recommending at this time that additional sediment samples be collected from Standley Lake for isotopic analyses and dating. We will review the EG&G Rocky Flats data when it is available to help direct our design.

It is also desirable to collect information from other lakes in the region that may have been impacted by the RFP. Sediments collected from such lakes could help determine how far the plumes extended. Unlike soil, deep sediment that is continuously submerged is not subject to erosion. Thus, any plutonium deposited from the air would remain in place. Some data have been collected from other bodies of water, most notable Calkins Lake, Boulder Reservoir, Boyd Lake, Autry Reservoir, Cherry Creek Reservoir, Marston Lake, Ralston Reservoir, Halligan Reservoir and Wellington Lake (Poet and Martell 1972, EPA 1971, and EPA 1973). However, the majority of data were obtained from surface samples only. For this reason, we propose sampling sediment from select lakes, dating the cores using ^{137}Cs and ^{210}Pb, and analyzing the samples for plutonium isotopes.

Cohen et al. 1990 provided the most detailed analysis of sediments collected from background lakes (Wellington Lake and Halligan Reservoir). Core samples were collected and analyzed for ^{239}Pu and ^{137}Cs. Lead-210 was used to date the core segments. The results were compared with those from Great Western Reservoir. This study indicated the need to collect more baseline data from other lakes for comparison with RFP impoundments. However, we are not recommending any additional background study at this time. The determination of specific isotopes of plutonium in sediment samples can be used to accurately identify the source of the plutonium (i.e., RFP versus fallout) in sediment.

Air

As discussed in the December 1993 progress report (Rope et al. 1993), the RFP contractor did not begin plutonium-specific analyses of routine air samples until around 1970. Prior to that time, the only routinely collected data available are total long-lived alpha (TTLα) concentrations. These non-specific counts are subject to interpretation problems (compared to plutonium-specific analyses), because of higher detection levels and difficulty in separation of site-released alpha emitters from naturally occurring alpha-emitters. Because of their importance to reconstruction of highest releases from the RFP, we are re-examining the airborne TTL*a* concentrations for Phase II of the dose reconstruction. There were studies, which should allow us to relate TTLα concentration to plutonium concentrations in ambient air samples (with some uncertainty). However, if archived air filters had been retained, it would be preferable to analyze them for plutonium and americium directly.

However, from interviews with past RFP employees, we are not optimistic that the RFP retained their ambient air filters from the time of the 1957 and 1969 fires or the periods of highest releases from the 903 Pad. Werkema (1980) states that in 1978, air filters were only analyzed specifically for plutonium if the TTLα count was more than 0.009 pCi m^{-3}. All others were stored for 18 months for possible plutonium analysis.

A number of documents have been located which describe studies investigating data quality issues for ambient air sampling. These issues include the air sampler collection efficiency,

efficiency of filter collection media, effect of sampling height above the ground, particle size issues (including natural ambient aerosols), and analytical methods. Although there will undoubtedly be some missing information, what has been located appears adequate to judge the quality and uncertainty of the past data. We do not think any additional monitoring would provide important new information to assess the quality of past air monitoring data. Therefore, no additional air monitoring studies are being recommended.

Vegetation

With the exception of special monitoring conducted in association with the 1957 and 1969 fires, specific plutonium data are not available for vegetation samples prior to 1970 (Rope et al. 1993). Routine off site vegetation monitoring for the years 1963 to 1970 consists of gross alpha analyses of samples. Preoperational vegetation samples collected in 1951 and initial monitoring program samples collected in 1952 and 1953 were also analyzed for gross alpha. The gross alpha data for vegetation samples collected between 1952 and 1970 may be useful for assessing spatial and temporal trends—and possibly for verifying routine release estimates. However, if archived vegetation samples had been retained, it would be preferable to analyze them for plutonium and americium directly. Based on interviews with past RFP employees, we are not optimistic that the RFP retained these samples.

Data Needs Identified during Conduct of Tasks 2 and 3

In Phase II, we are focusing on the following source terms: (1) plutonium released during the 1957 fire, (2) plutonium released from the 1969 fire, (3) routine plutonium releases, (4) historical releases of plutonium from the 903 Pad area, (5) carbon tetrachloride, (6) beryllium, and (7) plutonium and tritium in surface water. Data needs for completing Tasks 2 and 3 are presented below.

Plutonium Released from 1957 and 1969 Fires

It is possible to quantify, or at least identify, plutonium releases from episodic events through the analysis of downwind sediment cores. This is addressed in greater detail in the previous discussion on sediment.

One additional study that has been suggested is to obtain the waste materials from the 1957 and 1969 fires and analyze them for plutonium. The purpose of such a study would be to confirm the estimates for the plutonium recovered after the fires. The waste materials were shipped to the Idaho National Environmental Laboratory for disposal. The wastes from the 1957 fire were buried in the Subsurface Disposal Area of the Radioactive Waste Management Complex (RWMC) and are irretrievable. The wastes from the 1969 fire were emplaced on asphalt pads in the Transuranic Storage Area of the RWMC. The waste is theoretically retrievable; however, it is not realistically retrievable within the time frame of the dose reconstruction. The barrels of waste were emplaced, along with other barrels, in layers, separated by plywood, and covered with soil. Thus, it is not possible to conduct the proposed study.

Routine Plutonium Releases

Because the majority of plutonium in environmental media, such as soil and sediment, was deposited as a result of non-routine releases, particularly the 903 Pad release, it is doubtful that new samples would provide any additional information on routine plutonium releases. As such, no new studies are proposed.

Carbon Tetrachloride and Beryllium

It is highly unlikely that carbon tetrachloride persists in the environment. Therefore, no studies involving measurement of this contaminant are proposed.

Unlike in the case of plutonium, it is not possible to distinguish the source of the beryllium in the environment (i.e., from the RFP or other industries). Moreover, monitoring data indicate that beryllium concentrations in the RFP environment are indistinguishable from background. Therefore no new studies are proposed for measuring beryllium in environmental media.

It has been suggested that the archived tissue samples used in the study by Cobb et al. (1982) could be analyzed for beryllium to help evaluate human exposure to this metal. However, because it would be impossible to determine the source of beryllium, the information gained from such a study would not be useful to the dose reconstruction. Therefore, it is not recommended that these tissues be analyzed for beryllium.

Following a presentation given on berylliosis cases by Kathleen Kriess at the May HAP meeting, it was suggested that a beryllium sensitivity study could be conducted on individuals in the region around the RFP. The purpose of such a study would be to determine what fraction of the population are sensitized (generally thought to be less than 1% of the population) and could be susceptible for berylliosis. However, the sensitization is not necessarily a marker for exposure and cannot be related to the magnitude of beryllium released to the environment. For this reason, we do not recommend pursuing this study.

Plutonium and Tritium In Surface Water

Tritium from historical releases is no longer measurable in aquatic media. Plutonium has been shown to deposit in discernible patterns in the sediments of impoundments in the Rocky Flats environment. For these reasons, there is little need to initiate new studies to measure contaminants in surface waters near the RFP.

903 Pad Area

In 1969, the 903 former oil drum storage area was paved and designated the 903 Pad area. Oil drums stored in the 903 area prior to removal and paving had corroded and leaked, releasing plutonium contaminated oil to the soil. During drum removal and paving operations, plutonium contaminated soil was suspended and deposited in the field east of the pad. The field adjacent to and directly east of the Pad has been studied since then as a source of airborne plutonium. According to Langer (1991), the following processes have been considered:

- Saltation (wind erosion of bare soil with large particles impacting and suspending smaller particles)

- Wind resuspension of particles from grass blades
- Rain splash
- Mechanical disturbances and grass fires.

Total resuspension has been found to be low, estimated at 200 µCi/yr. Most of the material being resuspended from the 903 field is associated with soil and grass litter particles, with airborne radioactivity roughly proportional to the mass of particles collected.

Langer discussed the studies that have been performed to measure resuspension in the 903 field. Past studies involving reflection from lasers, and capture of saltating particles, failed to demonstrate significant resuspension, even at high wind speeds. He ascribes these observations to the soil becoming crusty over time, and to protection of soils by the grass canopy. Langer explains apparent contradictions with earlier work by Sehmel and others showing significant resuspension, by noting 903 field disturbance (ditch construction) prior to Sehmel's pre-1972 measurements. Langer states that it took 9 months for the effects of this disturbance to disappear. He also describes tests demonstrating that, for grass-covered areas, resuspension from grass is, over all, one to two orders of magnitude less significant that resuspension from open, dry, resuspendable soil.

Given the relatively significant release of plutonium from the 903 Pad, understanding resuspension is obviously important to the dose reconstruction. We have previously recommended that resuspension studies be pursued. However, three new studies, which are related to resuspension may provide information for the dose reconstruction have been funded for FY 95. The studies are to be conducted by EG&G Rocky Flats and are briefly described as follows:

> *Upwind-Downwind Sampling*: Air samplers will be set up "windward" and "downwind" of a known contaminated surface area. These two samplers will be linked by computer to measure concentrations only when the wind blows between the two samplers (within a given sector). The purpose of this experiment is to distinguish between airborne concentrations that are a result of upwind sources or "background" and airborne concentrations that are due only to resuspension.
>
> *Release of Pu from Vegetation*: Wind tunnel experiments similar to the type performed for OU3 operable unit (Standley Reservoir, Great Western Reservoir), will be performed on vegetated plots contaminated with Pu. This study is a follow-on study to the work Langer did in 1991. The purpose of this study is to examine the release of Pu from vegetation.
>
> *Portable Solar Powered Air Sampler*: A portable solar powered trailer containing air monitoring equipment will be designed and field tested. This trailer can then be set up at sites where remediation activities are taking place to monitor airborne particulate and activity concentrations. The equipment is planned to be set up at various sites where different operations are taking place such as installing a power pole or grading a road. In this way, releases due to specific construction activities may be quantified.

The first two of these studies are applicable to dose reconstruction while the third study has limited applicability because its focus is on the development of a measurement tool. The second study (release of Pu from vegetation), is probably the most important as far as providing information to dose reconstruction. Quantifying these resuspension rates as a function of wind speed and other conditions (precipitation, season) would be valuable information in terms of modeling releases from the field east of the 903 pad. In the past studies, Langer measured resuspension rates from vegetation (Langer 1991) and found no correlation between these rates and wind speed or direction. Sehmel (Sehmel 1980) also observed a non-correlation between wind speed and Pu release rates. Langer's observations were attributed to the poor sampling statistics of the study. A new, more detailed study using a wind tunnel would provide the necessary control to establish any relationship between wind speed and resuspension rates. The first study (Upwind-Downwind) will also provide valuable information for determining release rates from contaminated surface soil.

A limited amount of data is available concerning the actual quantity of plutonium that contaminated the soil in the 903 Pad area. Most estimates of the quantity of ^{239}Pu released from the area rely on concentrations measured in soil downwind of the site. One possible problem with this approach is that the material resuspends so that the amount in surface soil may not be strongly related to the original amount deposited. Also, unlike in sediment, deposits on soil are not datable. The dating and analysis of undisturbed sediment downwind of the 903 Pad area, as proposed previously in this report, may help quantify deposition rates during the period of concern.

It has also been suggested that nearby landfills might provide temporal information. Landfills are generally covered with a layer of soil daily. That soil may be a good record of airborne plutonium concentrations for that day. Newspapers within each day's layer could provide accurate date information. Unfortunately, there are a lot of unknown factors associated with this type of study. For example, newspapers may not be intact during the period of interest. Soil layers may become mixed due to redistribution by burrowing mammals. Leaching of plutonium may occur due to the presence of acids disposed of in the pit. These and other unquantifiable factors make this study unsuitable for dose reconstruction purposes. Sediment is a preferable medium for investigating depositional patterns over time as it has been demonstrated to provide useful information.

Summary of Data Objectives and Potential Studies

Table 5 presents a summary of all potential studies and data objectives mentioned in the previous sections. The studies are presented in the following general order: air, soil, sediment, vegetation, resuspension studies, and human exposure studies. The table includes a brief description of each study, major data objectives, historical does calculation component, and specific data needed. It includes all studies addressed, even if the study is considered to be unnecessary or unfeasible. The rationale for excluding or including a specific study in the final list of recommended studies is illustrated in Figure 3, presented previously in this report.

SELECTON OF RECOMMENDED STUDIES

The general procedure for excluding or including a specific study in the final list of recommended studies is illustrated in Figure 3, presented previously in this report. Table 6 presents the final recommendations. The specific rationale for including or excluding each study is summarized in Table 6. The study numbers refer to those assigned in Table 5.

In summary the final recommendations are

1. Pursue studies involving the collection and analysis of sediment cores from Standley Lake and other area lakes (to be selected based on history and suitability) for plutonium isotopes, ^{137}Cs and ^{210}Pb

2. Continue CSU urine bioassay study

3. Analyze archived air filters and vegetation samples, if they can be found.

In Part 2 of Task 5, study designs will be developed, with input from monitoring experts and HAP members. The final report will thus detail specific recommendations for conducting each study.

Table 5. Summary of Potential Studies and Data Objectives

Potential study	Major data objective[a]					Dose calculation component[b]						Specific data needed
	1	2	3	4	5	A	B	C	D	E	F	
1. Analysis of archived effluent air samples for Pu and Am	×					×						Direct measurements of airborne releases from RFP
2. Analysis of archived ambient air samples from stations downwind of 903 Pad area and from other ambient stations for Pu and Am				×					×	×		Pu concentrations in air to validate dispersion model or to compare with soil concentrations to derive deposition velocities and resuspension rates
3. Analysis of archived soil samples for Pu and Am		×		×			×			×		Results of reanalysis of past soil studies to confirm results used in developing source terms and model validation work
4. Collection and analysis of soil samples around the RFP for Be	×		×				×					Concentrations in the environment to verify offsite releases from RFP
5. Analyze more soil samples (additional and archived) for ^{239}Pu and ^{240}Pu isotopes			×			×						Isotopic ratios to distinguish source of Pu (RFP or fallout) and establish background levels
6. Collection and analysis of soil samples downslope of 903 Pad for ^{239}Pu and ^{240}Pu								×				Vertical and horizontal concentration patterns in soil downslope of 903 Pad area to determine pathways. Isotopic ratios to identify source of Pu.
7. In situ simulated rainfall and lab study of downward leaching of Pu in soil				×				×				Data on transport of Pu in soil and availability for surface erosion
8. Collection and analysis of sediment core samples from Great Western Reservoir, Standley Lake and other downwind lakes for Pu, ^{137}Cs, and ^{210}Pb	×		×	×		×	×	×				Temporal record in sediment core segments to identify major release events (via air and water) and pathways to lakes, verify source terms, and/or validate deposition estimates
9. Collection and analysis of sediment core samples from background lakes for Pu, ^{137}Cs, and ^{210}Pb	×					×	×					Baseline data to distinguish contamination from RFP releases from fallout
10. Collection and analysis of sediment core samples from Standley Lake, Great Western Reservoir, and background impoundment for Be and CCL_4	×					×						Temporal record in sediment core segments to help estimate and/or verify potential offsite releases from RFP
11. Analysis of cores collected from landfills around the RFP for Pu and ^{137}Cs	×					×	×			×		Temporal record to help estimate and/or verify 903 Pad and other releases

Table 5. Summary of Potential Studies and Data Objectives

Potential study	Major data objective[a]					Dose calculation component[b]						Specific data needed
	1	2	3	4	5	A	B	C	D	E	F	
12. Collection and analysis of woody vegetation cores for Pu			×		×		×					Temporal record of Pu in tree rings to identify historical airborne release events
13. Collection and analysis of lichens for Pu and Am		×			×			×				Record of Pu and Am in lichens to provide a spatial record of deposition/resuspension and verify soil results
14. Analysis of archived vegetation samples for Pu and Am	×		×	×						×		Concentrations of Pu in vegetation following specific events to validate release estimates or deposition calculations
15. Mechanical resuspension studies				×					×			Empirical resuspension rates to model 903 Pad area releases
16. Natural resuspension studies				×					×			Empirical resuspension rates to model resuspension of contaminated soil
17. Continue CSU urine bioassay study for Pu				×					×		×	Pu body burdens to validate integrated exposure estimates
18. Analysis of unclaimed crematoria ashes for Pu				×							×	Data on total body burdens from different periods of time
19. Analysis of archived human organs (from Cobb study) for Be				×					×		×	Be concentrations in organs to document human exposure to Be
20. Be sensitivity study				×							×	Determine sensitivity of human population for Be disease
21. Reanalysis of waste from cleanup after 1957 and 1969 fires	×				×		×					Confirm estimates of plutonium remaining after fire

[a] 1 = verify and/or refine existing source term; 2 = verify past monitoring results and clarify associated uncertainties; 3 = provide confirmation of past offsite contamination levels; 4 = provide new data for model development, calibration, or validation; and 5 = address any critical public concerns.

[b] A = source term development; B = source term verification; C = model structure; D = parameter estimation; E = model validation; and F = exposure assessment.

Table 6. Final Recommendations

Potential study[a]	Decision	Rationale for decision
1	Accept, if possible	Air filters for the period of interest have not been located and have probably been destroyed. However, in the unlikely event that they are found, we will analyze them as proposed.
2	Accept, if possible	Air filters for the period of interest have not been located and have probably been destroyed. However, in the unlikely event that they are found, we will analyze them as proposed.
3	Reject	Jones et al. (1994) demonstrated statistically, using data collected from 1970 to 1991 by CDH, that there is no evidence of a time trend in Pu concentrations in surface soil. Any differences in sample depth. Thus, it is possible to compare historical data with current data to confirm historical measurements and associated estimates of source terms.
4	Reject	Not possible to distinguish RFP Be from other sources of Be. Also, monitoring data indicate no difference between on site and off site concentrations.
5	Being done	Current investigations of background levels will be adequate for does reconstruction purposes.
6	Reject	Little new insight would be gained due to the high variability in Pu concentrations in soil near the 903 Pad. Recent EG&G studies, using simulated rainfall and pits, probably will provide sufficient data to elucidate soil pathways. Isotopic ratios not needed because of high levels of Rocky Flats Pu, compared to background concentrations, at this location.
7	Being done	EG&G Rocky Flats is doing this.
8	Accept (Standley Lake and other potentially impacted lakes only)	Standley Lake and downwind lakes (other than Great Western Reservoir) sediments should be studied. Much about airborne deposition can be learned from temporal trends in cores. Sediment is an excellent sink and, unlike soil, there is no loss other than decay. Methods proven by Hardy and Volchok (1980). Few cores taken on these lakes. Current EG&G study objectives may not be compatible with that of the does reconstruction. Great Western Reservoir should not be studied further because much data exist and recent CSU studies show that sediment layers have no discernible patterns, indicating mixing.
9	Reject	Current background data available. Also, if needed, isotopic ratios can be used to distinguish fallout Pu from RFP Pu in new samples.
10	Reject	$CC1_4$ unlikely to persist in sediments in measurable quantities. No possible to distinguish RFP Be from other sources of Be. Also environmental monitoring data indicate Be in sediments indistinguishable from background.
11	Reject	Unproven method. Too many unknowns: how to distinguish periods of time, transport factors (e.g., animals, leaching), etc.
12	Reject	Insufficient material available for analysis using conventional methods. Track etch method could possible be applied, but it is very expensive and unproven for vegetation. Small sample sizes limit the statistical analyses that can be used and, thus, the usefulness of the data. Too many unknown or unquantifiable factors contribute to concentrations in wood, sediment is better medium for evaluating temporal trends.

Table 6. Final Recommendations

Potential study[a]	Decision	Rationale for decision
13	Reject	Small sample sizes limit the statistical analyses that can be used and, thus, the usefulness of the data. Furthermore, it takes 30-40 years for a specimen to attain adequate sample mass for analysis. Therefore, can only sample in undisturbed areas, limiting value in assessing spatial patterns.
14	Accept, if possible	Archived vegetation samples have not been located and have probably been disposed of. However, in the unlikely event that they are found, we will analyze them as proposed.
15	Being done	EG&G Rocky Flats is doing this.
16	Being done	EG&G Rocky Flats is doing this.
17	Accept	Proven methods. Study is in place and just requires the collection of additional samples. May be the only way to measure integrated exposure using current population.
18	Reject	Lifestyles and location of individuals would be largely unknown making it difficult to link results with RFO releases.
19	Reject	Not possible to distinguish RFP Be from other sources of Be.
20	Reject	Would not reveal any information on RFP releases or impact on past populations.
21	Reject	Waste is irretrievable.

[a] Study numbers refer to those assigned in Table 5.

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